Growth of thin Fe/Fe_2O_3 films on the Cu(110) surface

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The growth of Fe/Fe-oxide double-layers on Cu(110) was studied with thermal energy atom scattering (TEAS), Auger electron spectroscopy, and low-energy electron diffraction (LEED). An iron film with a thickness of about 0.6 nm was evaporated at low temperature (130 K) on a smooth, well-ordered thin film of Fe_2O_3 prepared on Cu(110). This Fe film is disordered. Ordering of the film was observed at temperatures between 400 and 600 K. At 530 K, a structure corresponding to that of a well-ordered α-Fe(001) surface was observed with TEAS and LEED. Clear evidence was found for a mixing of the Fe and Fe_2O_3 layers at the interface, already beginning at the deposition temperature of 130 K. With increasing temperature, the mixing of the Fe and Fe_2O_3 layers became gradually more effective until, at around 600 K, it was essentially completed. Upon annealing the sample to 1000 K the structure of the film changes and a very thin (less than 2 ML) FeO film on top of the Cu(110) surface is obtained.
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I. INTRODUCTION

The growth of thin iron and iron oxide films have been studied extensively in the last decade, because of their interesting catalytic and magnetic behavior. For instance, spin valve structures[1] consisting of two very thin magnetic films separated by a nonmagnetic film (mostly Cu), are very promising materials for magnetic sensors and recording heads. With the ongoing increase in recording density, concepts of such spin valve structures are considered in which a Fe_2O_3 film is used as antiferromagnetic pinning layer.[2–5] An important aspect of spin valve structures based on the use of Fe_2O_3 is their thermal stability. Magnetic characterization of the thermal stability of spin valves composed of Fe_2O_3 as pinning layer in direct contact with a magnetic Co or iron alloy layer shows that around 500 K clear irreversible changes occur. However, a minor irreversible degradation of the magnetic properties seems to start already at 400 K and may be present even at room temperature.[2–4] Double-layer structures of Fe_2O_3 and pure Fe have also been investigated, but the thermal stability of these systems was not considered in detail.[6,7] For Fe/Fe–O multilayers, the layered structure was found to disappear completely above 570 K, whereas below this temperature only a limited decrease of the order of the sandwich structure was found.[8]

In the present work we report on experiments in which an ultrathin (0.6 nm), epitaxial Fe film is grown on a smooth, well-ordered Fe_2O_3 film prepared on a Cu(110) substrate. We used thermal energy atom scattering (TEAS) and low-energy electron diffraction (LEED) to characterize the structure and morphology of the surface. The composition of the film was characterized by Auger electron spectroscopy (AES). The preparation of the double layers is based on our recent investigation on the growth of epitaxial Fe_2O_3 on Cu(110):[9] a 5-ML-thick fcc γ-Fe film is prepared by evaporating Fe at 130 K, subsequent annealing of the film to 240 K, and freezing of the annealed sample to less than 150 K.[10] The surface of this film shows a structure which is identical to that of the Cu(110) surface. By oxidising the smooth γ-Fe(110) film at 130 K with 25 L O_2 and subsequent annealing of the sample to 700 K, a well-ordered iron oxide is formed which shows a (21×6) superstructure with respect to the substrate lattice. This superstructure is attributed to a coincidence lattice of the substrate and a slightly distorted (111) face of an ultra thin Fe_2O_3 film. At temperatures above 700 K, the Fe_2O_3 film is not stable. Upon annealing the sample to 1020 K another ordered oxide structure is found. The TEAS and LEED diffraction patterns measured after this preparation show a $(n\times8)$ superstructure with $n=17-19$, which was interpreted as resulting from a coincidence of the substrate lattice with the lattice of a slightly distorted FeO film.[9]

The thermal stability of the Fe films, deposited at 130 K on the Fe_2O_3/Cu(110) substrate, is investigated by slowly heating the sample and continuously monitoring the composition of the double layer by AES, and the order of the surface by TEAS. At several temperatures a more detailed structure analysis was performed by both TEAS and LEED.

II. EXPERIMENT

The experiments have been performed in an UHV-helium scattering apparatus at a base pressure of 5×10^{-11} mbar. The system is equipped with a supersonic helium nozzle beam with a quadrupole mass spectrometer as detector, a four-grid LEED optics, an AES spectrometer, and a quadrupole mass spectrometer for residual gas analysis. The apparatus has been described in more detail before.[11,12] The sample temperature T can be varied from 70 to 1200 K. By heating and cooling of the helium nozzle ($65\,\mathrm{K}<T_{\mathrm{nozzle}}<450\,\mathrm{K}$) the energy of the primary beam can be varied from 14 to 100 meV, corresponding to wavelengths between 1.21

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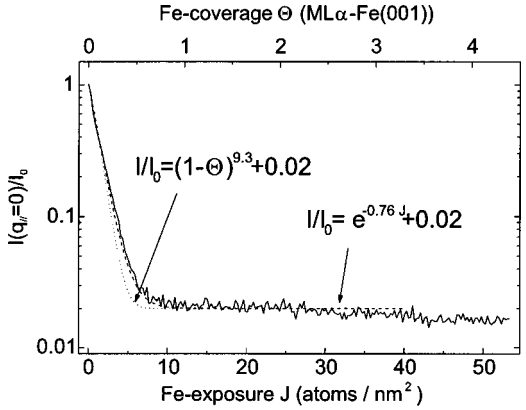

FIG. 1. Deposition of Fe on Fe_2O_3/Cu(110) at 130 K monitored with specular Helium scattering ($q_{\parallel}=0$) as a function of the Fe exposure J, the thickness of an ideal α-Fe film, respectively. The deposition rate is 0.01 ML/s. The He intensity is normalized to the intensity I_0, measured on the clean Fe_2O_3/Cu(110) surface at 130 K. The beam energy is 24.4 meV. The dashed line corresponds to an exponential decrease of the intensity.

and 0.45 Å. The He scattering measurements (TEAS) were performed at a total scattering angle of $\theta_t=\theta_i+\theta_f=90°$ where θ_i is the angle of incidence and θ_f the exit angle, both measured with respect to the surface normal. In the diffraction spectra the scattered He intensity is shown as a function of the in-plane component of the wave vector transfer $q_{\parallel}=|\mathbf{k}|(\sin\theta_i-\sin\theta_f)$, where $|\mathbf{k}|$ is the absolute value of the wave vector of the incoming atoms.

The Cu(110) sample was cut from a copper single crystal whose impurity concentration was less than 10^{-5}. The orientation of the surface was within 0.1° of the (110) direction. The sample was cleaned in the UHV chamber by repeated cycles of 800 eV Ar^+-ion bombardment followed by annealing at 800–900 K for several minutes. This procedure had to be repeated after each deposition experiment to remove the deposited Fe and Fe_2O_3. Iron was evaporated from a rod, 2 mm in diameter, by electron impact heating. The impurity concentration of the iron rod was less than 10^{-4}. The deposition rate could be kept constant within ±5% by monitoring the Fe-ion flux. During evaporation, the pressure in the main chamber did not rise measurably (less than 2×10^{-10} mbar). The calibration of the Fe evaporator was done both with TEAS and AES measurements as has been discussed in detail previously.[13]

III. RESULTS

A. Deposition of Fe on Fe_2O_3/Cu(110) at 130 K

An ordered Fe_2O_3 film on Cu(110) was prepared by oxidation (25 L O_2) at 130 K of a 5-ML-thick γ-Fe film, which was grown on a Cu(110) substrate, and by subsequent annealing of the oxide to 700 K. On the Fe_2O_3 film a thin Fe film was grown. The deposition of Fe on Fe_2O_3/Cu(110) at 130 K was monitored by specular He-scattering ($q_{\parallel}=0$) at a He-beam energy of 24.4 meV. In Fig. 1 the normalized specular intensity is shown as a function of the Fe exposure J, where the exposure is given both in atoms/nm^2 and in α-Fe(001) monolayers. The specular He intensity is found to decrease exponentially up to an exposure of about 6 Fe-atoms/nm^2 [~0.5 ML α-Fe(001)]. At higher exposures the specular intensity remains nearly constant. The constant scattering signal in this exposure range is attributed to Helium atoms which are incoherently scattered from a disordered surface. The intensity can be fitted by the function

$$\frac{I}{I_0}=A+e^{-CJ} \tag{1}$$

where $c=0.76$ (nm^2/Fe-atom) is a constant determining the slope of the curve in Fig. 1 and $A=0.02$ is the normalized intensity of the incoherently scattered background (dashed line in Fig. 1). The exponential decay can be explained by a process in which it is assumed that the Fe atoms adsorb randomly on the surface and that they form a disordered Fe layer at higher coverages. In that case, the specular He intensity is given by the equation[14]

$$\frac{I}{I_0}=A+(1-\Theta_1)^{\Sigma N_S} \tag{2}$$

with Σ being the cross section for incoherent scattering of a He atom from an adsorbed Fe atom, N_S the number of Fe atoms per unit surface area in the first layer at saturation, and Θ_1 the first layer coverage N_1/N_S where N_1 is the number of Fe atoms per unit area in the first layer. At low total coverage Θ all Fe atoms are in the first layer, i.e., $\Theta=\Theta_1$. With Θ substituted for Θ_1 in Eq. (2) the dotted curve in Fig. 1 is obtained. Though this curve describes the experimental data reasonable well, one does observe a discrepancy especially for $\Theta>0.2(I/I_0<0.15)$. Agreement in the whole exposure range of Fig. 1 is obtained if we assume ideal three-dimensional growth in which intralayer diffusion is forbidden. In that case one can, analogous to Langmuir adsorption, derive for the first layer Fe coverage Θ_1 the equation

$$\Theta_1=1-e^{-J/N_S}. \tag{3}$$

Inserting Eq. (3) in Eq. (2), one obtains Eq. (1) with $\Sigma=c=0.76\,\text{nm}^2$/Fe-atom. The observed crosssection seems to be reasonable in comparison to values which are usually found (typical values for Σ range from 0.3 and 2 nm^2).[15]

AES in the energy range below 100 eV is very sensitive to the chemical state of iron. Metallic iron and Fe^{3+} (Fe_2O_3), with AES transitions, respectively, at 47 and 52 eV can be clearly distinguished. In principle, Fe^{2+} (as in FeO) can also be distinguished from metallic iron, but the energy of the Fe^{2+} AES transition at 46.5 eV is too close to the AES transition of metallic Fe to resolve the two transitions in practical situations.[16–18] In Fig. 2, AES spectra are shown measured on γ-Fe(110)/Cu(110) [curve (a)], on Fe_2O_3(111)/Cu(110) [curve (b)], and on Fe/Fe_2O_3(111)/Cu(110) as prepared at 130 K [curve (c)]. In curve (a) measured on 5 ML γ-Fe (55 Fe-atoms/nm^2), two AES transitions are seen in the energy range below 70 eV: the 47 eV transition of metallic iron and the 61 eV transition of Cu. After oxidation of the γ-Fe film the AES spectrum has changed [curve (b)]. The 47 eV transition of metallic iron has disappeared and, instead, the 52 eV transition characteristic for Fe_2O_3 appears.[16–18] The Cu transition at 61 eV is still visible in curve (b), but it has decreased as a result of the oxidation of the Fe film, i.e., the Fe_2O_3 film is much thicker than the original Fe film, so the

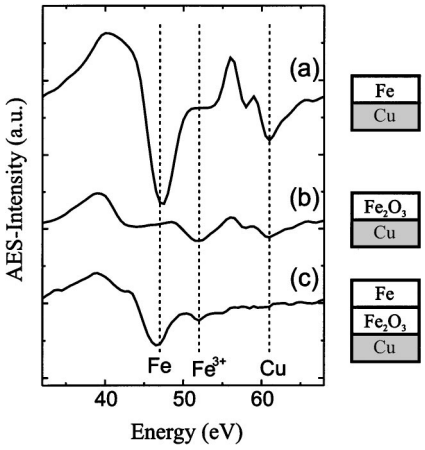

FIG. 2. AES spectra of 55 Fe-atoms/nm^2 [5 ML γ-Fe(110)] deposited on Cu(110) (a), of Fe_2O_3 on Cu(110) [5 ML γ-Fe on Cu(110) oxidized at 130 K with 25 L O_2 and subsequently annealed to 700 K] (b), and AES spectra of 55 Fe-atoms/nm^2 [4.5 ML α-Fe(001)] deposited on Fe_2O_3/Cu(110) (c). The AES transitions of metallic Fe (47 eV), Fe^{3+} corresponding to Fe_2O_3 (52 eV)and the AES transition of metallic Cu (61 eV) are marked with dashed lines. The primary energy of the electrons was 3 keV.

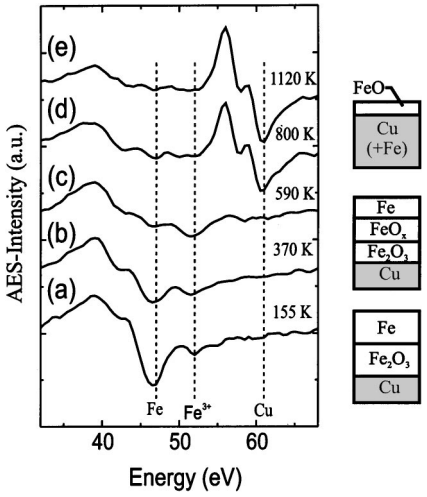

FIG. 3. AES spectra of 4.5 ML α-Fe deposited at 130 K on Fe_2O_3/Cu(110) measured during heating the surface with 4 K/min to 1170 K. Each spectrum is the average of 16 spectra. The average temperatures at which the spectra were measured is 155 (a), 370 (b), 590 (c), 800 (d), and 1120 K (e). The primary energy of the electrons was 3 keV.

copper is more effectively screened. Upon evaporation of 55 Fe atoms/nm^2 [4.5 ML α-Fe(001)] on Fe_2O_3(111)/Cu(110) the AES spectrum changes again [curve (c) of Fig. 2]. The Cu AES transition at 61 eV has almost disappeared due to the thick overlayer of Fe and Fe_2O_3. The transition at 52 eV, which is caused by Fe_2O_3, is still visible, but it has somewhat decreased, and the AES transition of metallic iron (47 eV) has appeared again, showing that there is a metallic iron film on top of the Fe_2O_3(111)/Cu(110) surface. The intensity of the 47 eV transition is a factor of 2 smaller in curve (c) than in curve (a) although about the same amount of iron was deposited in the two cases. We interpret this observation by mixing of the Fe and Fe_2O_3 films at the interface, i.e., some of the Fe atoms are oxidized by O atoms originating from the Fe_2O_3 layer. As discussed in the next section, we find strong evidence that this process already takes place in the temperature range between 130 and 200 K.

B. Annealing

After depositing 55 Fe atoms/nm^2 [4.5 ML α-Fe(001)] on Fe_2O_3(111)/Cu(110) at 130 K, the sample was heated continuously to 1170 K with a heating rate of 4 K/min. AES spectra were measured continuously with a rate of about one spectrum per 90 s. Figure 3 shows AES spectra between 30 and 70 eV, obtained by averaging 16 individual spectra. The spectra were measured at an average temperature of 155 (a), 370 (b), 590 (c), 800 (d), and 1120 K (e). Spectrum (a) shows AES transitions at 47 eV (metallic Fe) and at 52 eV (Fe^{3+}), reflecting the double-layer structure Fe/Fe_2O_3. At the Fe/Fe_2O_3 interface some Fe^{2+} may be present but that can not be resolved with our AES system. The Cu transition (61 eV) is not visible due to the thick overlayer of iron and iron oxide. During annealing two major changes in the AES spectra are observed. At low temperature ($T<600$ K), we found that the 47 eV AES transition (i.e., the minimum at this energy) disappears when the temperature increases, indicating that the thickness of the layer consisting of metallic Fe decreases. The 52 eV transition remains almost constant. However, a slight change of this transition could easily be masked by the change of the much larger transition at 47 eV. Cu is not diffusing to the surface in this temperature range as evidenced by the absence of the 61 eV transition, suggesting that there is essentially no intermixing between Cu and Fe below 600 K, i.e., all iron contained in the Fe and the Fe_2O_3 layer remains at the surface. This is confirmed by measurements of the 703 eV AES transition of iron.

At high temperature ($600\,\mathrm{K}<T<1170\,\mathrm{K}$) a further change of the AES spectra is observed. In the energy range between 42 and 52 eV, where the minima of the AES transitions of iron and iron oxide are expected, the AES spectrum is essentially without structure. Only the left shoulder of the Fe transition at 39 eV, which seems characteristic for all iron states,[16,17] is still visible [curves (d) and (e)]. In addition the AES transition at 61 eV is clearly observed. The AES spectra (d) and (e) in Fig. 3 are very similar to those measured on Cu(110) covered with a very thin FeO layer as was reported recently.[9] It should be remarked that the AES measurements can not exclude that Cu segregates on the FeO layer when the sample is heated to 1170 K. However, the structure analysis reported in the next section makes such an interpretation unlikely.

In Fig. 4 the temperature dependences of the 47 and 61 eV AES transitions are shown in more detail [Figs. 4(a) and 4(c)]. In addition, the figure shows the temperature dependence of the 511 eV (oxygen) AES transition [Fig. 4(b)] and the temperature dependence of the specular He intensity [Fig. 4(d)]. In the TEAS measurement the heating rate was 20 K/min, in the AES measurements 4 K/min. One should note that the AES signals in Fig. 4 are peak-to-peak amplitudes. For instance the peak-to-peak amplitude of the 47 eV transition is defined as $I(39\,\mathrm{eV})-I(47\,\mathrm{eV})$. As a consequence, this amplitude remains non zero at $T>600\,\mathrm{K}$ because the minimum of the AES intensity at 47 eV may have

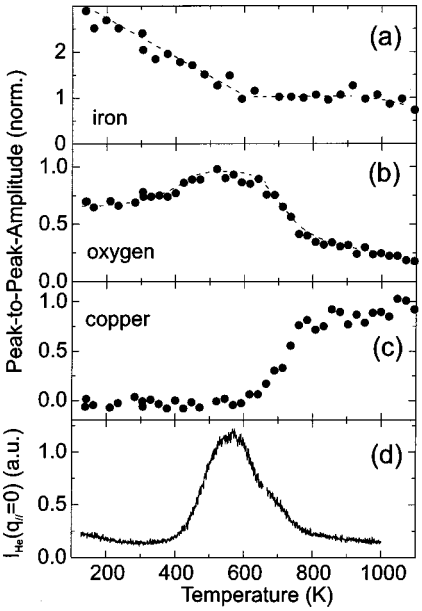

FIG. 4. Annealing of 4.5 ML α-Fe deposited on Fe_2O_3/Cu(110) at 130 K monitored with AES and TEAS. The AES measurements show the peak-to-peak amplitudes of the iron AES transition $[I(39\,\text{eV})-I(47\,\text{eV})]$ (a), the oxygen AES transition $[I(507\,\text{eV})-I(511\,\text{eV})]$ (b), and the Cu AES transition $[I(56\,\text{eV})-I(61\,\text{eV})]$ (c) during annealing. The TEAS measurement (e) shows the specular He intensity ($q_{\parallel}=0$) during annealing (beam energy $=24.4$ meV). The heating rate was 20 K/min in the case of the TEAS-measurement and 4 K/min in the case of the AES measurements.

disappeared, but the maximum at 39 eV is still present. The amplitudes of the 47 and the 511 eV transitions are normalized to the values measured on the $Fe_2O_3(111)/Cu(110)$ surface. The amplitude of the Cu transition (61 eV) is normalized to the amplitude measured at 1100 K.

The main effect observed at low temperature ($130\,\text{K} < T < 600\,\text{K}$) is a continuous change of the chemical composition of the Fe/Fe_2O_3 double layer: The peak-to-peak amplitude of the 47 eV AES transition (Fe) decreases linearly by a factor of about 3 [Fig. 4(a)]. The amplitude of the 511 eV AES transition (oxygen) increases, but only by a factor of 1.4, and the increase is not linear [Fig. 4(b)]. Such a behavior can be explained by a reaction between Fe and Fe_2O_3, first ($130\,\text{K} < T \leqslant 350\,\text{K}$) at the interface, resulting in a loss of metallic iron, but the oxygen remains located at roughly the same depth below the surface (no or little increase of the 511 eV signal is seen). As the temperature increases above 350 K, diffusion of oxygen atoms towards the surface becomes easier, i.e., a larger amount of the Fe film can be oxidized (reduction of the 47 eV signal) and screening of the oxygen AES signal by the iron layer is reduced (the 511 eV signal increases). At 600 K, the mixing seems to be completed in the sense that all the iron atoms in the top layer have reacted with oxygen as indicated by the 47 eV AES signal becoming constant at about this temperature. Ordering of the surface is found to occur between 400 and 600 K according to the increase of the specular Helium intensity in TEAS measurement shown in Fig. 4(d). The ordering and the diffusion of oxygen seem to be related processes as they occur in almost the same temperature range.

Above 600 K the double layer, which actually seems to be already an almost homogeneous FeO_x film, starts to disappear completely as evidenced by the decrease of the 511 eV AES amplitude (oxygen desorption) and the increase of the 61 eV AES signal (diffusion of Cu to the surface). The specular He intensity is found to decrease for $T > 600\,\text{K}$ which is mainly due to the Debye Waller attenuation [Fig. 4(d)]. There seems to be a hump in the curve around 700 K which might be due to a change of the surface structure, as discussed in the next section.

C. Surface structure

The structure of the $Fe/Fe_2O_3/Cu(110)$ surface annealed to 530, 730, and 1020 K was investigated with TEAS and LEED. The sample was heated with 20 K/min to the earlier mentioned temperatures and cooled down afterwards to the measurement temperature (100–120 K in case of LEED and 130 K in case of TEAS measurements). Figures 5(a) and 5(b) show Helium diffraction spectra of the $Fe_2O_3/Cu(110)$ substrate [curves (1)] and of the $Fe/Fe_2O_3/Cu(110)$ double layer annealed to 165 K [curves (2)], 530 K [curves (3)], 730 K [curves (4)], and 1020 K [curves (5)]. The intensity distributions of the He atoms measured in the [001] direction [Fig. 5(a)] and in the $[1\bar{1}0]$-direction [Fig. 5(b)] are shown as a function of the in-plane component of the wave vector transfer $q_{\parallel}$. Figure 6 shows LEED patterns of the $Fe_2O_3/Cu(110)$ substrate [Fig. 6(a)] and of a 0.6-nm-thick Fe film [4.5 ML α-Fe(001)] evaporated on $Fe_2O_3/Cu(110)$ at 130 K and subsequently annealed to 530 K [Fig. 6(b)] and 1020 K [Fig. 6(c)]. After annealing the sample up to 300 K, no LEED pattern could be observed. The LEED pattern observed after annealing the sample to 730 K was identical to the LEED pattern shown in Fig. 6(c) (1020 K). Also the LEED pattern for FeO/Cu(110) is shown [Fig. 6(d)], which we reported previously.[9]

The helium spectra, curves (1) in Figs. 5(a) and 5(b), and the LEED pattern of Fig. 6(a) are characteristic for the Cu(110) surface covered with an ultrathin Fe_2O_3 film (0.8–1.2 nm).[9] After deposition of 5 ML Fe on $Fe_2O_3/Cu(110)$ and heating the film to 165 K no diffraction pattern was observed with TEAS [Figs. 5(a) and 5(b), curves (2)], nor with LEED. This shows that the surface is disordered at low temperature. After annealing the film to 530 K [curves (3)], diffraction spots appear at $q_{\parallel}=0$, and $\pm(2.18\pm0.03)\,\text{Å}^{-1}$ in the [001]-direction and at $q_{\parallel}=-4.47\pm0.05$, -2.22 ± 0.02, 0, 2.19 ± 0.03, and $4.42\pm0.05\,\text{Å}^{-1}$ in the $[1\bar{1}0]$ direction. The position of the specular peak, which defines the zero of the $q_{\parallel}$ axis, can be determined with an accuracy of 0.002 Å^{-1}. The uncertainties given for the diffraction peaks are mainly due to an inaccuracy of the sample manipulator. We can assign the peaks to a zero, first and second order diffraction from a square lattice with a reciprocal lattice constant of $g^*=2.196\pm0.015\,\text{Å}^{-1}$ which corresponds to a lattice constant in real space of $g=2.86\pm0.02\,\text{Å}$. The LEED pattern measured on this surface shows diffraction spots arranged in a square [Fig. 6(b)]. Taking the LEED pattern of clean Cu(110) as reference, we find a reciprocal lattice constant $g^*=2.19\pm0.03\,\text{Å}^{-1}$ from Fig. 6(b), and thus a lattice con-

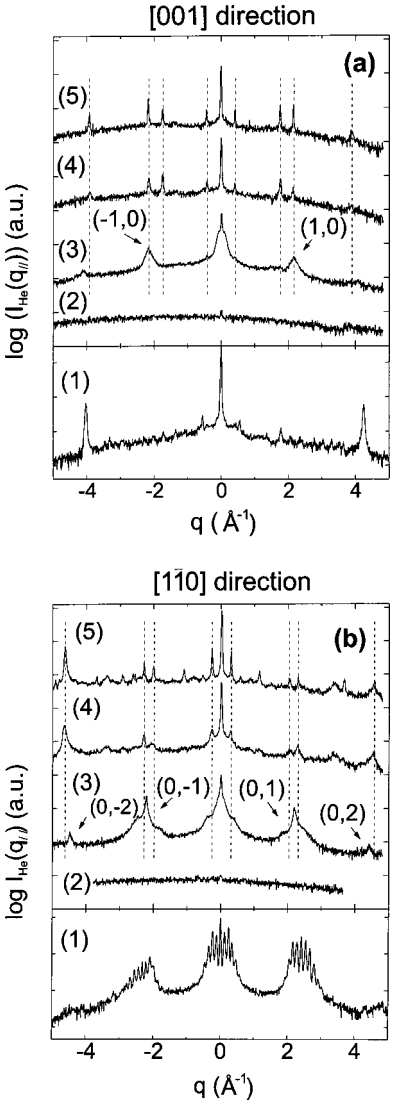

FIG. 5. TEAS spectra in the Cu-[001] direction (a) and in the Cu-[$1\bar{1}0$] direction (b) of Fe_2O_3/Cu(110) (1), and of a 55 atoms/nm^2 (4.5 ML α-Fe(001)−0.6-nm-thick) Fe film deposited at 130 K on Fe_2O_3/Cu(110) and subsequently annealed to 165 (2), 530 (3), 730 (4), and 1020 K (5), respectively. The energy of the He beam was 24.4 meV. The spectra were measured at 130 K.

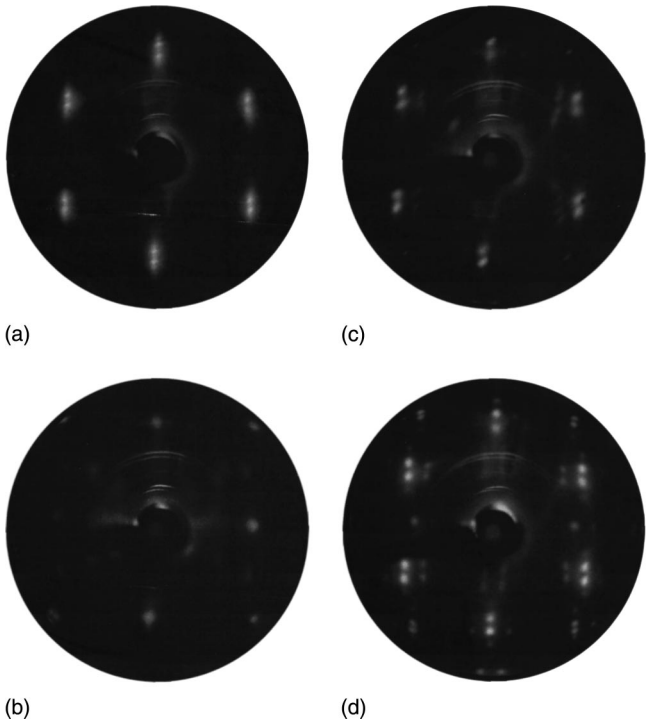

FIG. 6. LEED pattern obtained from Fe_2O_3/Cu(110) (a), 55 Fe-atoms/nm^2 [4.5 ML α-Fe(001)−0.6 nm] deposited on Fe_2O_3/Cu(110) at 130 K and subsequently annealed to 530 K (b), and 730 K (c), respectively, and LEED pattern from FeO/Cu(110) (d). The primary energy of the electrons was 70 eV. The patterns were measured at a temperature between 100 and 120 K.

stant of 2.87 ± 0.04 Å. These values agree very well with the attice constant of bcc α-Fe ($g = 2.87$ Å).[19] The smoothness of this surface can be estimated from the width of the specular beam [Figs. 5(a) and 5(b), curves (3)]. The full width of half maximum is $\Delta q_{\parallel} = 0.05$ Å^{-1} in both directions. The width of the coherently scattering area (domain) can then be estimated to be $2\pi/\Delta q_{\parallel} = 120$ Å.

In the LEED pattern [Fig. 6(b)] some additional spots can be observed which, however, are too weak to be analyzed in detail. With TEAS we observe rather broad first order diffraction peaks in the [001]direction [Fig. 5(a), curve (3)]. The specular peak in this direction seems to be a combination of a sharp peak superimposed on a much broader peak and two very small peaks are seen at $q_{\parallel} = \pm 0.4$ Å^{-1}. At about ± 4.1 Å^{-1}, two humps are seen which deviate significantly from second order diffraction from α-Fe(001). In the [$1\bar{1}0$] direction the broadening of the diffraction peaks is less prominent than in the [001] direction. The humps observed around them seem to develop in separate peaks.

When the sample is heated to temperatures larger than 530 K, the structure of the surface changes drastically. After heating to 730 K, TEAS spectra are observed [Figs. 5(a) and 5(b), curves (4)] in which the α-Fe(001) diffraction spots have disappeared and new diffraction peaks have appeared. The same spectra, but with more and sharper peaks, are observed after annealing the sample to 1020 K [Figs. 5(a) and 5(b), curves (5)]. It is interesting to note that the positions of the additional features observed in the TEAS spectra after heating the sample to 530 K [curves (3)], i.e., the small peaks observed at $q_{\parallel} = \pm 0.4$ Å^{-1} in the [001] direction and the humps at $q_{\parallel} = \pm 0.4$ Å^{-1} in the [$1\bar{1}0$] direction, correspond very well to peak positions in the spectra of the curves (4) and (5) in Figs. 5(a) and 5(b). Apparently, the transition of the surface structure from α-Fe(001) to the new structure is already taking place at 530 K. In fact, this is not surprising, because the AES measurements show that most of the iron has already reacted with oxygen when the surface is heated to 530 K. One may note that the small peaks seen at ± 4.1 Å^{-1} agree neither with the structure of the α-Fe(001) surface nor with the structure found at higher temperature [curves (4) and (5)].

The structure of the surface after heating to 1020 K can be identified with LEED. The observed diffraction pattern [Fig. 6(c)] is very similar to the one reported for diffraction from FeO/Cu(110) [Fig. 6(d)].[9] The TEAS spectra observed for FeO/Cu(110)[9] are somewhat different from those presented here, but the position of the peaks which are observed

here and in Ref. 9 correspond very well. In fact, a good reproducibility of the peak positions and yet a poor reproducibility of the actual spectra with respect to the intensities and visibility of several peaks was already found in our study of the FeO/Cu(110) surface. Interpretation of this high temperature structure by a thin FeO layer on top of the substrate (rather than a Cu layer segregated on FeO, see Sec. III B), is also supported by the observation that the transition to this structure essentially takes place between 530 and 730 K. Evidence for copper diffusing towards the surface is found only above 600 K, and even when heating with a five times slower rate only a limited increase of the Cu AES signal is found at 730 K [Fig. 4(c)]. We conclude therefore that the Fe/Fe_2O_3/Cu(110) surface transforms to a FeO/Cu(110) surface during heating to 1000 K.

IV. DISCUSSION AND SUMMARY

We have studied the surface structure and thermal stability of an ultrathin Fe/Fe_2O_3 double layer prepared on a Cu(110) substrate. Iron is found to adsorb randomly on the Fe_2O_3 surface upon evaporation at 130 K. The Fe film which is obtained after depositing 0.6 nm iron is disordered. We found evidence for a reaction between the newly deposited Fe film and the underlying Fe_2O_3 film beginning at or just above the deposition temperature. Probably the reaction takes place only at the interface between the Fe and the Fe_2O_3 film at these very low temperatures. If the oxide is oxygen terminated, 2 ML of the Fe layer (i.e., almost half of the metallic iron) can be oxidized requiring no or only little displacement of O atoms in the oxide. Upon annealing the sample, ordering of the surface of the Fe/Fe_2O_3 double layer is found to occur in the temperature range between 400 and 600 K. In about the same temperature range, oxygen atoms become more mobile giving rise to an increased mixing. The temperature dependence of the oxygen AES transition, when heating the double layer with 4 K/min, indicates that mixing of the oxygen and the iron is completed at 500 K. However, there is still metallic Fe left at this temperature as indicated by the 47 eV AES transition. This is confirmed by diffraction experiments (LEED, TEAS) which indicate that a metallic α-Fe(001) surface is present after heating the sample with 20 K/min to 530 K.[20] When heating the sample to temperatures larger than 600 K, the double-layer structure disintegrates completely: oxygen desorbs, Fe disappears probably into the bulk of the Cu sample, and a very thin FeO layer remains on the Cu(110) surface, which is stable up to 1100 K.

We have observed that, at 500 K, oxygen diffuses to the surface, but this process is slow at this temperature. Since we observe that most of the iron in the surface of a 0.6 nm film is still metallic at 500 K, we estimate that the mean diffusion length of the oxygen in the Fe film, on a time scale of minutes, is clearly smaller than 0.6 nm at this temperature. Accordingly, we expect that oxygen diffusion from the oxide in the Fe layer is negligible some 100 K below 500 K. Thus, we conclude that double-layers consisting of two or more nanometer Fe on Fe_2O_3 are essentially stable up to a temperature of about 400 K. Below this temperature only Fe atoms at or close to the interface seem to interact with oxygen from the Fe_2O_3 layer, and may become oxidized. However, without annealing to a higher temperature the surface of the double-layer remains disordered. Upon annealing to a temperature around 500 K for a few minutes, the double-layer becomes well ordered. For the more or less comparable Fe/Fe-O multilayer system, Ichikawa *et al.* reported a slow degradation when the multilayer is annealed at 570 K,[8] i.e., 40–70 K higher than the annealing temperature which we consider. However, the thickness of the Fe layers considered by Ichikawa *et al.* (4.6 nm) and thus the diffusion distance which is necessary to destroy the periodicity of the multilayer is much larger than the layer thickness which we consider (0.6 nm) and the longer annealing time (30 min instead of about 2 min) in their experiments cannot compensate completely for this difference. We conclude therefore that the stability of the Fe/Fe_2O_3 double layer is very similar to that of Fe/Fe–O double layers. With respect to spin valves using Fe_2O_3 as a pinning layer, one should consider oxygen diffusion from the Fe_2O_3 layer into the neighboring magnetic layer as possible origin for a deteriorating thermal stability at temperatures of 500 K or above. Annealing for a short period to a temperature of 500–600 K could, however, lead to better magnetic characteristics, because the improved ordering of the layers may outweigh the deterioration resulting from mixing of the layers. Indeed, both behaviors—improving and worsening of magnetic properties—have been found upon heating Fe_2O_3 containing spin valves to a temperature above 500 K.[3,4,21] However, it should be noted that these observations can not be compared in detail with the present results since they consider iron alloys rather than pure Fe as a magnetic layer in contact with the Fe_2O_3 pinning layer.

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